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Polarization of light scattered into second harmonic by free electrons

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Abstract. The polarization of the light scattered into second harmonic by free electrons, when the incident light is plane polarized, is discussed. The scattered light is also plane polarized in general. However, if the electric vector of the incident light is approximately in the plane of the paper (i.e., the plane containing the incident and the scattered beams), there is a sudden flip to perpendicular polarization of the scattered light at a critical angle, $\theta_c \approx 75{\cdot}5°$; however, the intensity of the scattered light then vanishes and the effect is unlikely to be observed.

It is shown that the second harmonic scattering cross section is largest when the scattering angle, θ, is approximately 90° and the incident light has the polarization angle $\phi_0 \approx 45°$; the polarization angle of the scattered light is then $\phi = 116{\cdot}5°$.

1. Introduction

The classical theory of the scattering of light into harmonics by free electrons has been given by one of us (Vachaspati 1962, 1963). If the incident light is plane polarized, the scattered light is also plane polarized. We calculate here the dependence of the angle of polarization of the scattered light on the polarization angle of the incident light and the angle of scattering. The results are discussed in the last two sections.

2. Definitions

We call the direction of the electric field as the direction of polarization. To choose appropriate coordinate systems for defining the polarization angles, we take a plane (the plane of the paper) containing the incident light and the observed light with unit vectors $\boldsymbol{n}_0$ and $\boldsymbol{n}$ respectively. Introduce a unit vector $\boldsymbol{\alpha}_1$ perpendicular to $\boldsymbol{n}$ in the plane of the paper and another unit vector $\boldsymbol{\alpha}_2$ perpendicular to the plane of the paper such that the unit vectors $(\boldsymbol{\alpha}_1, \boldsymbol{\alpha}_2, \boldsymbol{n})$ form a right-handed coordinate system. Explicitly,

$$\boldsymbol{\alpha}_1 = \frac{\boldsymbol{n}_0-(\boldsymbol{n}_0\cdot\boldsymbol{n})\boldsymbol{n}}{\sqrt{1-(\boldsymbol{n}_0\cdot\boldsymbol{n})^2}}, \qquad \boldsymbol{\alpha}_2 = \frac{\boldsymbol{n}\times\boldsymbol{n}_0}{\sqrt{1-(\boldsymbol{n}_0\cdot\boldsymbol{n})^2}}. \tag{1}$$

The angle which the electric vector of the scattered light makes with $\boldsymbol{\alpha}_1$ is called the angle of polarization of the scattered light and is denoted by φ; we do not distinguish between the angles φ and $\pi+\varphi$ and allow φ to range from 0 to π. A

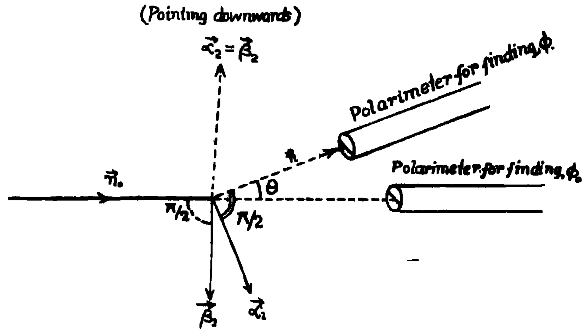

Figure 1. Electric vector of scattered light is resolved along mutually perpendicular unit vectors ($\boldsymbol{\alpha}_1$, $\boldsymbol{\alpha}_2$ and $\boldsymbol{n}$); that of the incident light is resolved along the unit vectors ($\boldsymbol{\beta}_1$, $\boldsymbol{\beta}_2$ and $\boldsymbol{n}_0$); $\boldsymbol{\alpha}_1$, $\boldsymbol{\beta}_1$, $\boldsymbol{n}_0$ and $\boldsymbol{n}$ are in the plane of the paper and $\boldsymbol{\alpha}_2 = \boldsymbol{\beta}_2$ is perpendicular to this plane pointing downwards.

similar coordinate system is defined for the incident light. The unit vectors $\boldsymbol{\beta}_1$, $\boldsymbol{\beta}_2$ and $\boldsymbol{n}_0$ form a right handed system; explicitly,

$$\boldsymbol{\beta}_1 = -\frac{\boldsymbol{n}-(\boldsymbol{n}_0\cdot\boldsymbol{n})\boldsymbol{n}_0}{\sqrt{1-(\boldsymbol{n}_0\cdot\boldsymbol{n})^2}}; \qquad \boldsymbol{\beta}_2 = \frac{\boldsymbol{n}\times\boldsymbol{n}_0}{\sqrt{1-(\boldsymbol{n}_0\cdot\boldsymbol{n})^2}} = \boldsymbol{\alpha}_2. \tag{2}$$

Notice the minus sign in $\boldsymbol{\beta}_1$; it has been inserted so that the directions $\boldsymbol{\alpha}_1$ and $\boldsymbol{\beta}_1$ coincide for forward scattering. The angle which the electric vector of the incident light makes with $\boldsymbol{\beta}_1$ is called the polarization angle of the incident light and is denoted by ϕ_0. Again, we do not distinguish between ϕ_0 and $\pi+\phi_0$ and let ϕ_0 range from 0 to π. (It is shown later that ϕ_0 needs to be considered between 0 and $\pi/2$ only, see Discussion C). The angle of scattering is denoted by θ and varies from 0 to π.

3. Electric Field

The incident beam is taken as plane polarized; its electric field is

$$\boldsymbol{E} = E_0\boldsymbol{e}_0 \cos(k_0(x_0-\boldsymbol{n}_0\cdot\boldsymbol{x})); \tag{3}$$

$$(\boldsymbol{e}_0\cdot\boldsymbol{\beta}_1) = \cos\varphi_0; \quad (\boldsymbol{e}_0\cdot\boldsymbol{\beta}_2) = \sin\varphi_0. \tag{4}$$

The electric field of the scattered light can be obtained from the expressions given by Vachaspati (1963) it is

$$\boldsymbol{E}^{Scatt} = \frac{e}{r}\{\boldsymbol{M}-(\boldsymbol{n}\cdot\boldsymbol{M})\boldsymbol{n}\}.$$

For the second harmonic harmonic light, $\boldsymbol{M}$ can be replaced by

$$\boldsymbol{N}^{(2)}\sin 2\psi_0,$$

where

$$\boldsymbol{N}^{(2)} = C^{(2)}\boldsymbol{e}_0+D^{(2)}\boldsymbol{n}_0;$$

$$C^{(2)} = -2k_0q\cos\alpha;\quad D^{(2)} = -\tfrac{1}{2}k_0q;$$

$$q = \frac{e^2E_0^{\,2}}{m^2k_0^{\,2}};\ \cos\alpha = (\boldsymbol{n}\cdot\boldsymbol{e}_0);\ \psi_0 = k_0(x_0-|\boldsymbol{x}|);$$

(Note that the angle α is not to be confused with the unit vectors $\boldsymbol{\alpha}_1$ and $\boldsymbol{\alpha}_2$ of equation (1)).

so that

$$\boldsymbol{E}^{Scatt} = \frac{e}{r}[C^{(2)}\boldsymbol{e}_0+D^{(2)}\boldsymbol{n}_0-\{C^{(2)}(\boldsymbol{n}\cdot\boldsymbol{e}_0)+D^{(2)}(\boldsymbol{n}\cdot\boldsymbol{n}_0)\}\boldsymbol{n}]\sin 2\psi_0. \tag{5}$$

Notice that the electric vector, $\boldsymbol{E}^{Scatt}$, is transverse to $\boldsymbol{n}$:

$$\boldsymbol{E}^{Scatt}\cdot\boldsymbol{n} = 0$$

as it should be.

4. Polarization

We resolve $\boldsymbol{e}_0$ and $\boldsymbol{n}_0$ along the three axes provided by $\boldsymbol{\alpha}_1$, $\boldsymbol{\alpha}_2$ and $\boldsymbol{n}$. The result is

$$\boldsymbol{E}^{Scatt} = \frac{e}{r}A(\cos\varphi\,\boldsymbol{\alpha}_1+\sin\varphi\,\boldsymbol{\alpha}_2)\sin 2\psi_0, \tag{6}$$

where

$$A\cos\varphi = C^{(2)}\cos\theta\cos\varphi_0+D^{(2)}\sin\theta, \tag{7a}$$

$$A\sin\varphi = C^{(2)}\sin\varphi_0. \tag{7b}$$

If we keep our polarimeter so as to receive the incident light, we can find φ_0 by measuring the angle which the electric vector makes with $\boldsymbol{\beta}_1$ in the plane $(\boldsymbol{\beta}_1, \boldsymbol{\beta}_2)$; the angle φ is measured when the instrument is receiving the scattered light and the angle is in the $(\boldsymbol{\alpha}_1, \boldsymbol{\alpha}_2)$ plane.

When the values of $C^{(2)}$ and $D^{(2)}$ given above are used, we find

$$\cos\varphi = \frac{2\cos\theta\cos^2\phi_0-\frac{1}{2}}{\sqrt{(2\cos\theta\cos^2\phi_0-\frac{1}{2})^2+\sin^2 2\phi_0}}; \tag{8a}$$

$$\sin\varphi = \frac{\sin 2\varphi_0}{\sqrt{(2\cos\theta\cos^2\varphi_0-\frac{1}{2})^2+\sin^2 2\varphi_0}}; \tag{8b}$$

and

$$\tan\varphi = \frac{\sin 2\varphi_0}{2\cos\theta\cos^2\varphi_0-\frac{1}{2}}; \quad 0 \leqslant \phi < \pi. \tag{9}$$

As we do not wish to distinguish between ϕ and $\phi+\pi$, the use of (9) is preferable to that of (8a, b).

5. Discussion

From (9), we notice the following :

(A) If $\phi_0 = 0$, then $\phi = 0$ for all θ. This means that, if the incident light is polarized in the plane of the paper, the scattered light is also polarized in the plane of the paper, no matter what the angle of scattering is. The case of $\phi_0 = 0$ and $\theta = \cos^{-1} 1/4 \approx 75{\cdot}5°$ is, however, exceptional and is discussed in D below.

(B) If $\phi_0 = \pi/2$, then, again, $\phi = 0$ for all θ. This means that, if the incident light is polarized perpendicular to the plane of the paper, the scattered light is polarized in the plane of paper, no matter what the angle of scattering is.

(C) From (9), we see that the right hand side is unaltered if we replace ϕ_0 by $\pi-\phi_0$. That is, the polarization of the scattered light is the same for initial polarization ϕ_0 and $\pi-\phi_0$. Therefore, we need consider ϕ_0 as ranging from 0 to $\pi/2$ only.

(D) (i) If the polarization of the incident light is such that

$$\cos\phi_0 = \tfrac{1}{2}\sqrt{\sec\theta},$$

then $\phi = \pi/2$; i.e., the scattered light is polarized perpendicular to the plane of the paper. This relation is possible only if

$$1/4 \operatorname{Sec}\theta < 1,$$

$$\text{i.e.}\quad 0 \leqslant \theta < \theta_c, \quad \theta_c = \cos^{-1} 1/4 \approx 75{\cdot}5°.$$

For angles of scattering greater than the critical angle θ_c, the scattered light cannot be ploarized perpendicular to the plane of the paper. For angles of scattering,

θ, less than 75·5°, the polarization of the scattered light can be perpendicular to the plane of the paper if ϕ_0 and θ are appropriately chosen from Figure 2. It shows that, for each initial polarization ϕ_0, we can find an angle of scattering θ at which the light is polarized perpendicular to the plane of the paper. The case $\phi_0 = 0$ needs more discussion which is done below in D (ii).

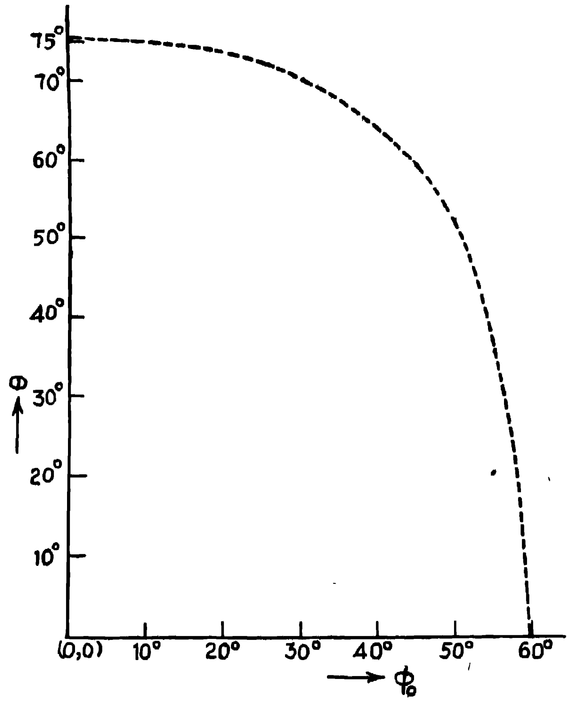

Figure 2. For certain angles of polarization of the incident light, the scattered light is polarized perpendicular to the plane of the paper ($\phi = \pi/2$) at particular angles of observation, θ. The relation between the incident polarization angle and the corresponding scattering angle, for which $\phi = \pi/2$, is plotted here.

(D) (ii) We get some interesting results in case the initial light is polarized in the plane of the paper and the angle of observation is the critical angle, θ_c. In (A) we saw that, for $\phi_0 = 0$, the scattered light is always polarized in the plane of the paper; it would therefore seem that the polarization suddenly changes from 0 to $\pi/2$ at $\theta_c = 75·5°$ and the scattered light is thus suddenly polarized perpendicularly to the plane of the paper as soon as we get to the observation angle θ_c; but, if we deviate from this angle ever so slightly, we would find the polarization to be in the plane of the paper. To see whether this is true, we take ϕ_0 as slightly different from 0 and explore the polarization at an angle of scattering, θ, which differs from θ_c by $\delta\theta$:

$$\theta = \theta_c + \delta\theta.$$

One can then easily see that (9) becomes

$$\tan\phi = -\frac{4}{\sqrt{15}}\frac{\varphi_0}{\delta\theta}.$$

This expression can assume any value depending on what ϕ_0 and $\delta\theta$ are. In practice, the finite aperture of the observing apparatus will have the effect that, for the observed scattered beam, $\delta\theta$ may range from, say, $-1°$ to $+1°$ (see Figure 3. If the incident radiation is not polarized exactly in the plane of the paper, the scattered radiation will then contain polarizations ranging from almost $\phi = 0$ to $\phi = \pi/2$, the observer will find the radiation depolarized. Thus, for $\phi_0 \approx 0$, the scattered light gets depolarized at $\theta = \theta_c \approx 75{\cdot}5°$.

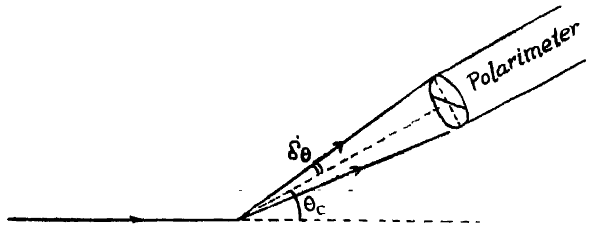

Figure 3. The figure shows that, on account of the finite aperture of the observing apparatus, it is not possible to observe the scattered light at exactly the critical angle θ_c, but one must allow a width $\pm\delta\theta$.

(E) The dependence of the angle of polarization of the scattered light on the incident light polarization and the scattering angle is shown in Figure 4.

(F) The scattering cross section for the second harmonic is given as

$$\left[\frac{d\sigma^{(2)}}{d\Omega}\right]_{\text{Polarized light}} = \frac{1}{4}\left(\frac{e^2}{m}\right)^2 q[\sin^2\theta + 4\sin^2 2\alpha - 8\cos^2\alpha\cos\theta]$$

which can be written in terms of ϕ_0 and θ as

$$\left[\frac{d\sigma^{(2)}}{d\Omega}\right]_{\text{Polarised light}} = \frac{1}{4}\left(\frac{e^2}{m}\right)^2 q \sin^2\theta$$

$$\times[1+16\cos^2\phi_0(1-\sin^2\theta\cos^2\phi_0)-8\cos^2\phi_0\cos\theta].$$

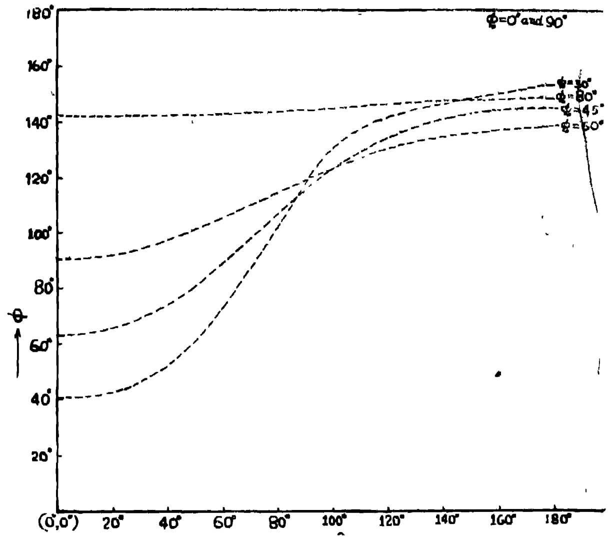

Figure 4. Polarization of the scattered light, ϕ, plotted against the angle of scattering, θ, for various incident polarizations ϕ_0. Since ϕ and $\pi+\phi$ represent the same polarization, the curves for $\phi_0 = 0$ and $\phi_0 = \pi/2$ coincide. Moreover, the curves for ϕ_0 and $\pi-\phi_0$ are the same. For $\phi_0 \approx 0$, $\theta = \theta_c \approx 75{\cdot}5°$ represents a critical point where there is a sudden flip in the polarization of the scattered light.

The dependence of this cross section on ϕ_0 and θ can be seen from Figures 5 and 6. It will be seen that the cross section is the largest near $\phi_0 \approx 45°$ and $\theta = 90°$. For these values of ϕ_0 and θ, the scattered light has the polarization angle $\phi \approx 116{\cdot}5°$.

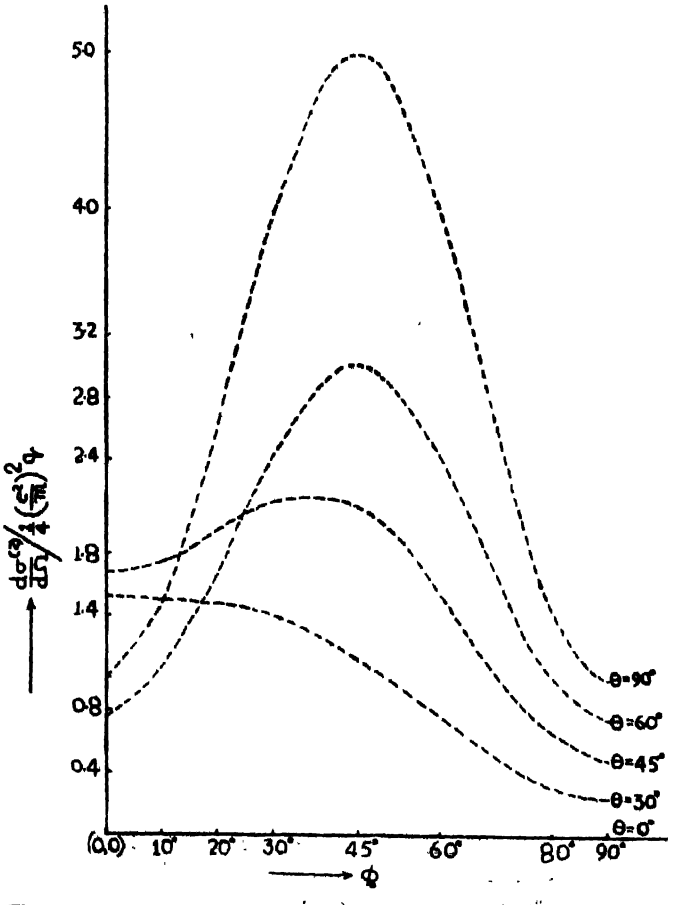

Figure 5. The second harmonic scattering cross section is plotted here against the initial polarization, ϕ_0, for various angles of observation. Notice that the cross section is largest at $\phi_0 \approx 45°$.

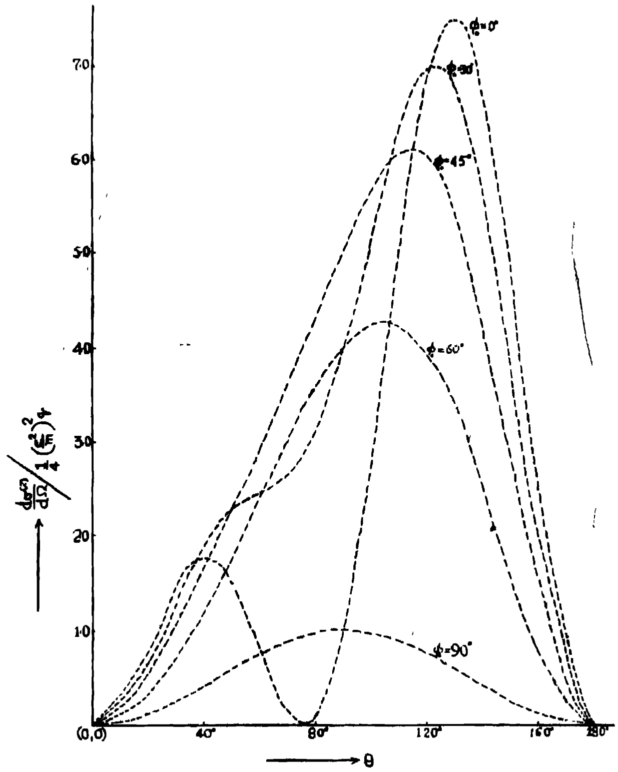

Figure 6. The second harmonic cross section is plotted here against the angle of scattering for various initial polarization. Notice that the cross-section is largest at $\theta \approx 90°$. It should be observed that the cross section vanishes for $\phi_0 = 0$, $\theta = \theta_0 \approx 75 \cdot 5°$.

6. Concluding Remarks

From the above discussion, it is clear that the most interesting case would be the one when the incident light is polarized approximately in the plane of the paper and the scattered light is observed at various scattering angles, for all scattering angles except one, the second harmonic scattered light will be plano polarized in the plane of the paper; there will be a sudden polarization flip at the critical angle, $\theta_c = 75{\cdot}5°$. Unfortunately the second harmonic scattering cross section also vanishes at this angle; therefore, it may not be possible to observe the polarization flip. So far as the ease of experiments is concerned, it is greatest near $\phi_0 \approx 45°$ where the cross section is large (Figure 6); the polarization of the second harmonic scattered light will then follow the curve for 45° in figure 5.

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References

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